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(54) **ORGANIC ELECTROLUMINESCENT ELEMENT**

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H01L 51/00 (2006.01)

C09K 11/06 (2006.01)

(52) **H05B 33/14** (2006.01)

U.S. Cl.

CPC **H01L 51/0072** (2013.01); **C09K 11/06**

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2211/1092 (2013.01); **C09K 2211/185**

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H01L 51/0088 (2013.01)

(58) **Field of Classification Search**

None

See application file for complete search history.

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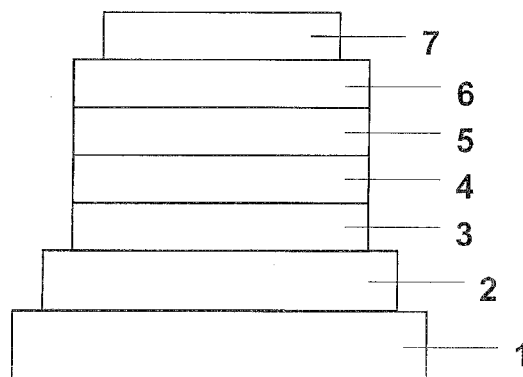
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Primary Examiner — Gregory Clark

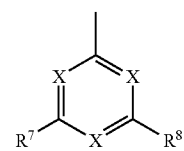
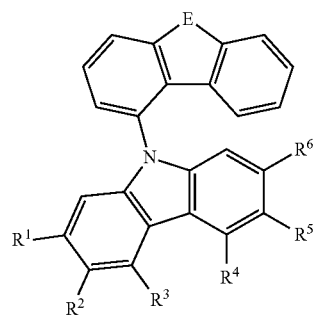
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(57) **ABSTRACT**

Provided is an organic electroluminescent device (organic EL device) which has improved luminous efficiency and a simple configuration, while ensuring sufficient driving stability. This organic electroluminescent device includes a light-emitting layer between an anode and a cathode that are laminated on a substrate. The light-emitting layer contains a phosphorescent light-emitting dopant, and a carbazole compound represented by the following formula (1) as a host material. In the formula (1), E represents oxygen or sulfur, and R¹ to R⁶ each represent hydrogen, an alkyl group, a cycloalkyl group, or an aromatic group represented by the formula (2). In the formula (2), X represents CR⁹ or nitrogen.



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7 Claims, 2 Drawing Sheets

Fig.1

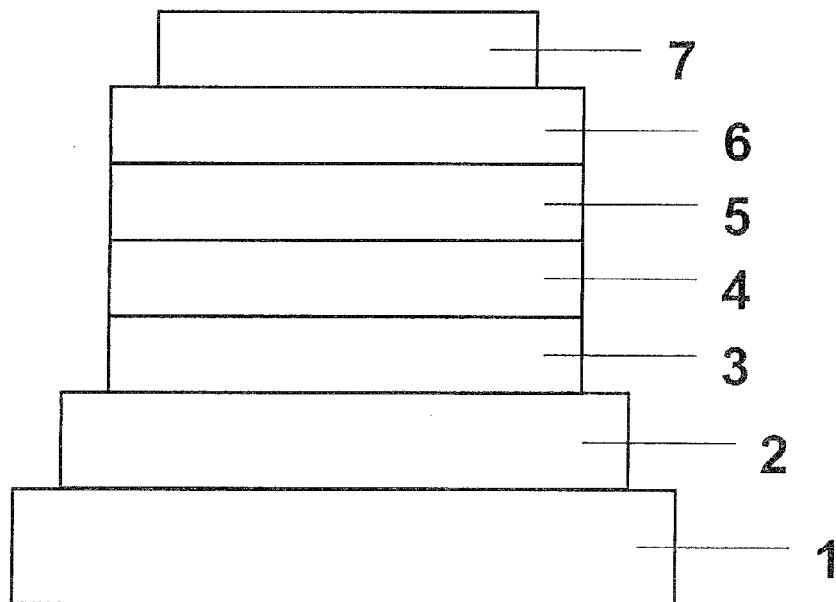


Fig.2

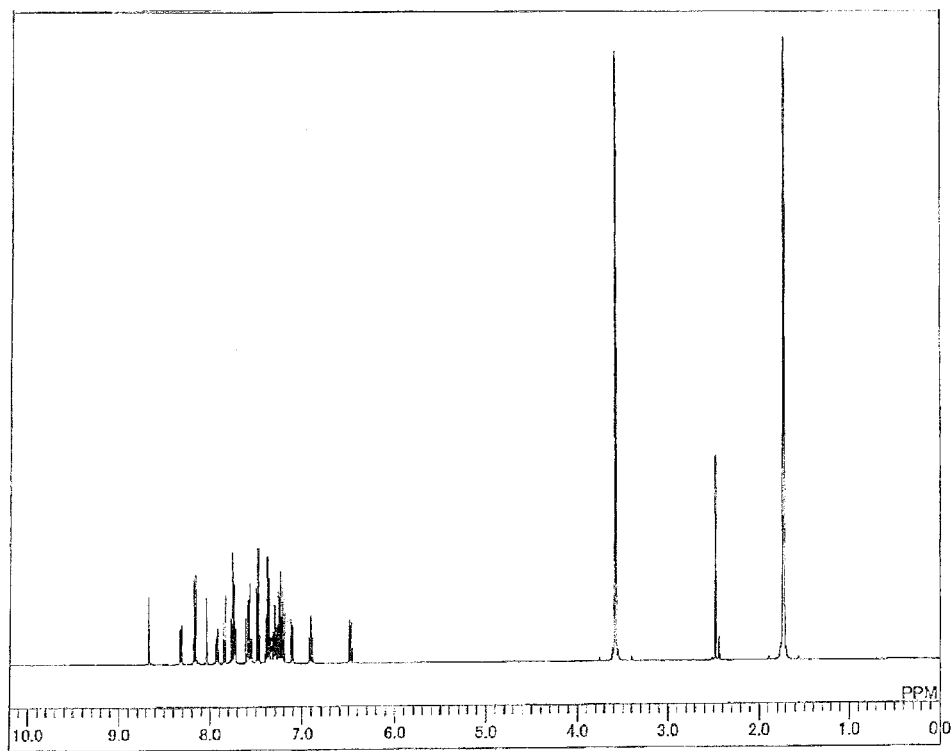
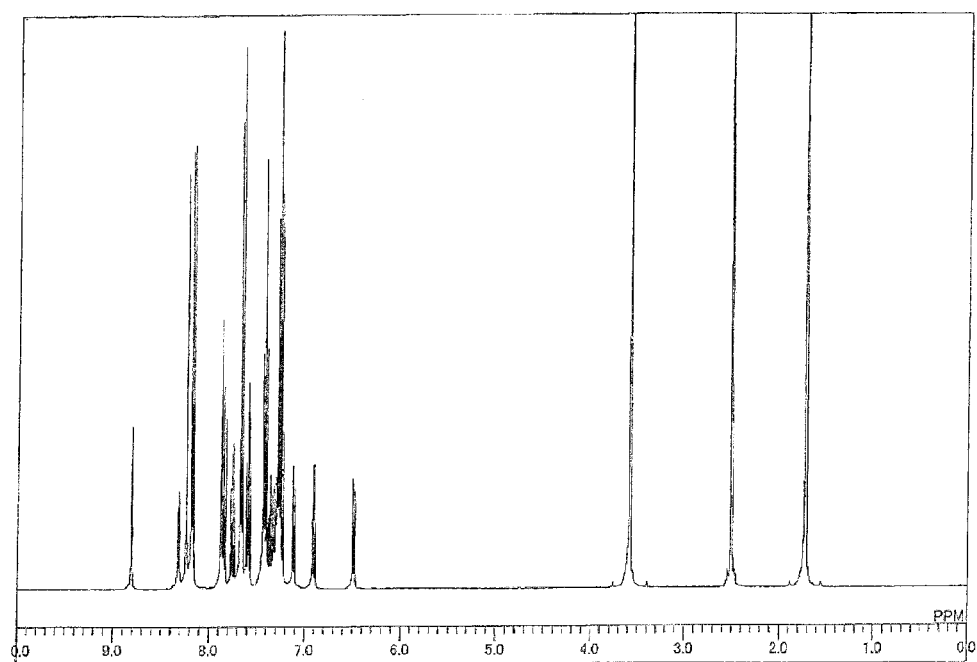


Fig.3



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ORGANIC ELECTROLUMINESCENT
ELEMENT

TECHNICAL FIELD

The present invention relates to an organic electroluminescent device using a novel material for an organic electroluminescent device, and specifically, to a thin-film-type device that emits light when an electric field is applied to a light-emitting layer formed of an organic compound.

BACKGROUND ART

In general, an organic electroluminescent device (hereinafter referred to as organic EL device) is constructed of a light-emitting layer and a pair of counter electrodes interposing the light-emitting layer therebetween in its simplest structure. That is, the organic EL device uses the phenomenon that, when an electric field is applied between both the electrodes, electrons are injected from a cathode and holes are injected from an anode, and each electron and each hole recombine in the light-emitting layer to emit light.

In recent years, progress has been made in developing an organic EL device using an organic thin film. In order to enhance luminous efficiency particularly, optimization of kinds of electrodes has been attempted for the purpose of improving efficiency of injection of carriers from the electrodes. As a result, there has been developed a device in which a hole-transporting layer formed of an aromatic diamine and a light-emitting layer formed of an 8-hydroxyquinoline aluminum complex (hereinafter referred to as Alq3) are formed between electrodes as thin films, resulting in a significant improvement in luminous efficiency, as compared to conventional devices in which a single crystal of anthracene molecules or the like is used. Thus, development of the above-mentioned organic EL device has been promoted in order to accomplish its practical application to a high-performance flat panel having features such as self-luminescence and rapid response.

Further, studies have been made on using phosphorescent light rather than fluorescent light as an attempt to raise luminous efficiency of a device. Many kinds of devices including the above-mentioned device in which a hole-transporting layer formed of an aromatic diamine and a light-emitting layer formed of Alq3 are formed emit light by using fluorescent light emission. However, by using phosphorescent light emission, that is, by using light emission from a triplet excited state, luminous efficiency is expected to be improved by about three times to four times, as compared to the case of using conventional devices in which fluorescent light (singlet) is used. In order to accomplish this purpose, studies have been made on adopting a coumarin derivative or a benzophenone derivative as a light-emitting layer, but extremely low luminance has only been provided. Further, studies have been made on using a europium complex as an attempt to use a triplet state, but highly efficient light emission has not been accomplished. In recent years, many studies centered on an organic metal complex such as an iridium complex have been made, as disclosed in Patent Literature 1, for the purpose of attaining high luminous efficiency and a long lifetime.

CITATION LIST

Patent Literature

[PTL 1] JP 2003-515897 A
[PTL 2] JP 2001-313178 A

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[PTL 3] WO 2009/008100

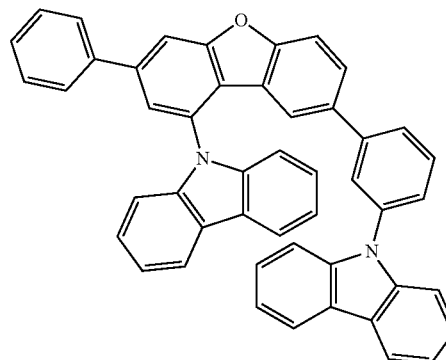
[PTL 4] JP 2011-509247 A

[PTL 5] WO 2011/057706

Not only the dopant material but also a host material to be used is important for obtaining high luminous efficiency. A typical material that has been proposed as the host material is, for example, 4,4'-bis(9-carbazolyl)biphenyl (hereinafter referred to as "CBP") as a carbazole compound introduced in Patent Literature 2. When CBP is used as a host material for a green phosphorescent light-emitting material typified by a tris(2-phenylpyridine)iridium complex (hereinafter referred to as "Ir(ppy)3"), owing to the characteristic of CBP by which the flow of a hole is facilitated and the flow of an electron is made difficult, a charge injection balance is broken and excessive holes flow out to an electron-transporting layer side. As a result, the efficiency of light emission from Ir(ppy)3 reduces.

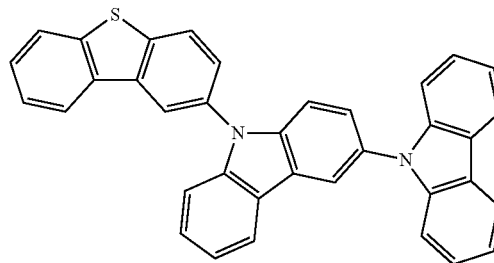
As described in the foregoing, a host material having a high triplet excitation energy and balanced injecting/transporting characteristics for both charges (a hole and an electron) is needed for obtaining high luminous efficiency in an organic EL device. Further, a compound that is electrochemically stable, and has high heat resistance and excellent amorphous stability has been desired, and hence an additional improvement has been required.

Patent Literature 3 discloses such a carbazole compound as shown below as a host material for an organic EL device.



However, it is assumed that the carbazole derivative does not provide sufficient luminous efficiency because the derivative has a phenyl group at each of the 3- and 7-positions of dibenzofuran.

Patent Literature 4 discloses such a carbazole compound as shown below as a host material for an organic EL device.

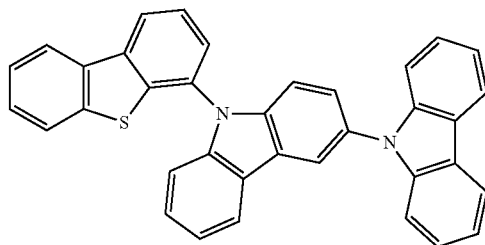


However, the disclosed compound is merely a compound obtained by introducing carbazole to the 2-position of dibenzothiophene, and the literature does not disclose the useful-

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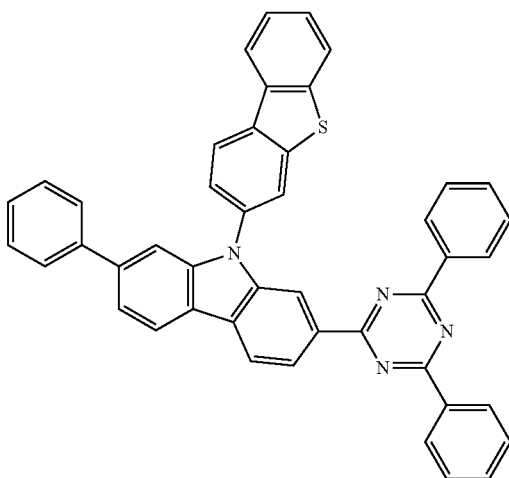
ness of an organic EL device using the compound of the present invention obtained by introducing carbazole to the 1-position of dibenzothiophene or dibenzofuran.

Patent Literature 4 discloses such a carbazole compound as shown below as a host material for an organic EL device.



However, the disclosed compound is merely a compound obtained by introducing carbazole to the 4-position of dibenzothiophene, and the literature does not disclose the usefulness of an organic EL device using a compound obtained by introducing carbazole to the 1-position of dibenzothiophene or dibenzofuran.

Patent Literature 5 discloses such a carbazole compound as shown below as a host material for an organic EL device.



The disclosed compound is characterized in that a nitrogen-containing six-membered ring and carbazole are introduced, and the 3-position of dibenzothiophene is substituted in the compound. However, the literature does not disclose the usefulness of an organic EL device using a compound obtained by introducing carbazole to the 1-position of dibenzothiophene or dibenzofuran.

SUMMARY OF INVENTION

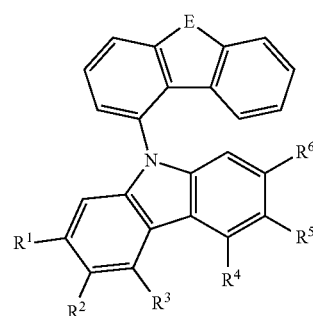
In order to apply an organic EL device to a display device in a flat panel display or the like, it is necessary to improve the luminous efficiency of the device and also to ensure sufficiently the stability in driving the device. The present invention has an object to provide, in view of the above-mentioned circumstances, an organic EL device, which has high efficiency, has high driving stability, and is practically useful and a compound suitable for the organic EL device.

As a result of their extensive studies, the inventors of the present invention have found that the use of a compound,

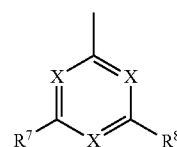
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which has a carbazole group at the 1-position of dibenzothiophene or dibenzofuran and has a specific substituent at any one of the 2- to 7-positions of the carbazole group, in an organic EL device causes the device to show excellent characteristics. Thus, the inventors have completed the present invention.

The present invention relates to an organic electroluminescent device, including: a substrate; an anode; an organic layer; and a cathode, the anode, the organic layer, and the cathode being laminated on the substrate, in which a carbazole compound represented by the general formula (1) is used in at least one layer selected from the group consisting of a light-emitting layer, a hole-transporting layer, an electron-transporting layer, a hole-blocking layer, and an electron-blocking layer.



(1)



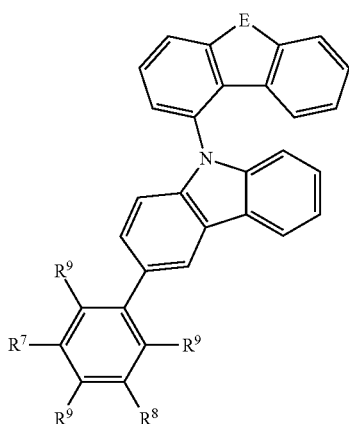
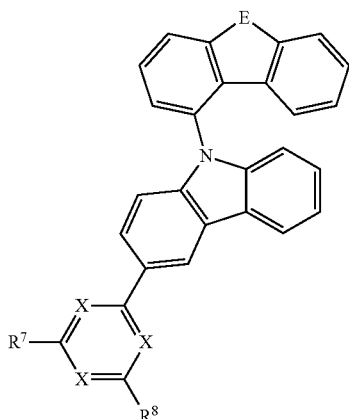
(2)

In the general formula (1), R^1 to R^6 each independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, or an aromatic group represented by the general formula (2), and at least one of R^1 to R^6 represents an aromatic group represented by the general formula (2), X's each independently represent CR^9 or nitrogen, R^7 to R^9 each independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, an aromatic hydrocarbon group having 6 to 18 carbon atoms, or an aromatic heterocyclic group having 3 to 17 carbon atoms, and the aromatic hydrocarbon group and the aromatic heterocyclic group may each have a substituent, and E represents oxygen or sulfur.

In the general formula (1), at least one of R^1 to R^6 represents a monovalent aromatic group represented by the general formula (2). It is preferred that R^2 represent an aromatic group represented by the general formula (2).

In addition, the carbazole compound represented by the general formula (1) is preferably a carbazole compound represented by the general formula (3), more preferably a carbazole compound represented by the general formula (4).

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In the general formulae (1) to (4), the same symbols have the same meanings. Therefore, the meanings of X, R⁷ to R⁹, and E are described in the general formulae (1) and (2). It should be noted that in the general formula (4), a plurality of R⁹'s may be identical to or different from one another.

Although in the general formula (4), R⁷ to R⁹ are as described above, it is preferred that R⁷ to R⁹ each independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, an unsubstituted aromatic hydrocarbon group having 6 to 18 carbon atoms, or an unsubstituted aromatic heterocyclic group having 3 to 17 carbon atoms, and it is also preferred that R⁷ to R⁹ each independently represent a substituted aromatic hydrocarbon group or substituted aromatic heterocyclic group obtained by providing any such aromatic hydrocarbon group or aromatic heterocyclic group with a substituent. The substituent is preferably an alkyl group having 1 to 6 carbon atoms, a cycloalkyl group having 3 to 10 carbon atoms, an alkoxy group having 1 to 6 carbon atoms, an acyl group having 2 to 7 carbon atoms, an aromatic hydrocarbon group having 6 to 12 carbon atoms, or a diarylamino group having 12 to 24 carbon atoms. In addition, the aromatic heterocyclic group is preferably an unsubstituted aromatic heterocyclic group having 3 to 17 carbon atoms that is not a nitrogen-containing six-membered ring, and the aromatic heterocyclic group can have a substituent. In the calculation of the number of carbon atoms, in the case of a substituted aromatic hydrocarbon group or a substituted aromatic heterocyclic group, the number of carbon atoms of a substituent is excluded. However, the number

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including the number of carbon atoms of the substituent preferably falls within the range.

In addition, the organic electroluminescent device of the present invention preferably includes a light-emitting layer containing the carbazole compound represented by the general formula (1) and a phosphorescent light-emitting dopant.

BRIEF DESCRIPTION OF DRAWINGS

FIG. 1 is a sectional view illustrating a structural example of an organic EL device.

FIG. 2 shows a ¹H-NMR chart of a carbazole compound 8.

FIG. 3 shows a ¹H-NMR chart of a carbazole compound 49.

DESCRIPTION OF EMBODIMENTS

An organic electroluminescent device of the present invention contains a carbazole compound represented by the general formula (1) in a specific layer. A carbazole compound represented by the general formula (3) or (4) is available as the carbazole compound represented by the general formula (1). In the general formula (1), at least one of R¹ to R⁶ represents an aromatic group represented by the general formula (2).

In the general formula (1), R¹ to R⁶ each independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, or a monovalent aromatic group represented by the general formula (2), and at least one of R¹ to R⁶ represents an aromatic group represented by the general formula (2). R² preferably represents an aromatic group represented by the general formula (2).

In the general formulae (2) and (3), X's each independently represent CR⁹ or nitrogen. In addition, in the general formulae (2) to (4), R⁷ to R⁹ each independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, an aromatic hydrocarbon group having 6 to 18 carbon atoms, or an aromatic heterocyclic group having 3 to 17 carbon atoms. Here, the aromatic hydrocarbon group having 6 to 18 carbon atoms or the aromatic heterocyclic group having 3 to 17 carbon atoms can have a substituent.

When any one of R¹ to R⁹ represents an alkyl group having 1 to 10 carbon atoms or a cycloalkyl group having 3 to 11 carbon atoms, specific examples thereof include a methyl group, an ethyl group, a propyl group, a butyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, a cyclohexyl group, and a methylcyclohexyl group, and the group may be linear or branched. An alkyl group having 1 to 6 carbon atoms or a cycloalkyl group having 3 to 8 carbon atoms is preferred. Specific examples thereof include a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, and a cyclohexyl group.

Next, the case where any one of R⁷ to R⁹ represents an aromatic hydrocarbon group or an aromatic heterocyclic group is described.

The aromatic hydrocarbon group or the aromatic heterocyclic group is an aromatic hydrocarbon group having 6 to 18 carbon atoms or an aromatic heterocyclic group having 3 to 17 carbon atoms. Specific example thereof include monovalent groups obtained by removing one hydrogen atom from an aromatic compound selected from benzene, naphthalene, fluorene, anthracene, phenanthrene, fluoranthene, pyrene, chrysene, pyridine, pyrimidine, triazine, indole, qui-

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noline, isoquinoline, quinoxaline, naphthyridine, carbazole, acridine, phenanthroline, phenazine, benzofuran, dibenzofuran, xanthene, oxanthrene, phenoxazine, benzothiophene, dibenzothiophene, thioxanthene, thianthrene, phenoxathiin, and phenothiazine. Of those, monovalent groups obtained by removing one hydrogen atom from an aromatic compound selected from the following compounds are preferred: benzene, indole, carbazole, benzofuran, dibenzofuran, benzothiophene, and dibenzothiophene. Monovalent groups obtained by removing one hydrogen atom from an aromatic compound selected from the following compounds are more preferred: benzene and carbazole.

The aromatic hydrocarbon group or the aromatic heterocyclic group may have a substituent, and when any such group has a substituent, the total number of substituents is 1 to 10, preferably 1 to 6, more preferably 1 to 4. In addition, when the aromatic hydrocarbon group or the aromatic heterocyclic group has 2 or more substituents, the substituents may be identical to or different from each other. In addition, in the calculation of the number of carbon atoms of the aromatic hydrocarbon group or the aromatic heterocyclic group, when any such group has a substituent, the number of carbon atoms of the substituent is not included. When the calculation is performed while the number of carbon atoms of a substituent is included, in the case of an aromatic hydrocarbon group, the number of carbon atoms falls within the range of preferably 6 to 50, more preferably 6 to 30. In the case of an aromatic heterocyclic group, the number of carbon atoms falls within the range of preferably 3 to 50, more preferably 5 to 30.

Preferred examples of the substituent include an alkyl group having 1 to 6 carbon atoms, a cycloalkyl group having 3 to 10 carbon atoms, an alkoxy group having 1 to 6 carbon atoms, an acyl group having 2 to 7 carbon atoms, an aromatic hydrocarbon group having 6 to 12 carbon atoms, and a diarylamino group having 12 to 24 carbon atoms. More preferred examples thereof include an alkyl group having 1 to 4 carbon atoms, a cycloalkyl group having 3 to 8 carbon atoms, an alkoxy group having 1 to 4 carbon atoms, an acyl group having 2 to 5 carbon atoms, an aromatic hydrocarbon group having 6 to 10 carbon atoms, and a diarylamino group having 12 to 20 carbon atoms, and specific examples of such groups can include a methyl group, an ethyl group, a propyl group, a butyl group, a cyclopentyl group, a cyclohexyl group, a methylcyclohexyl group, a methoxy group, an ethoxy group, a propoxy group, a butoxy group, a hexyloxy group, an acetyl group, a propionyl group, a phenyl group, a tolyl group, a naphthyl group, a diphenylamino group, and a dinaphthylamino group.

The compound represented by the general formula (1) is preferably represented by the general formula (3) or (4). In each of the general formulae (1), (3), and (4), E represents oxygen or sulfur. In each of the general formulae (3) and (4), R⁷ to R⁹ are as described above.

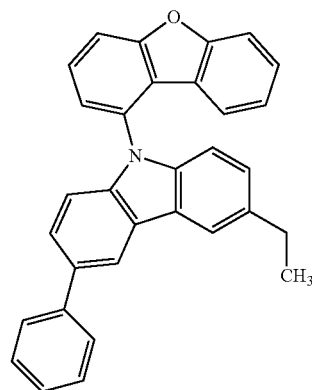
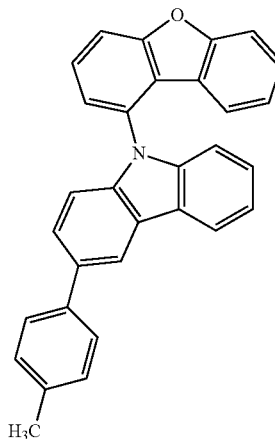
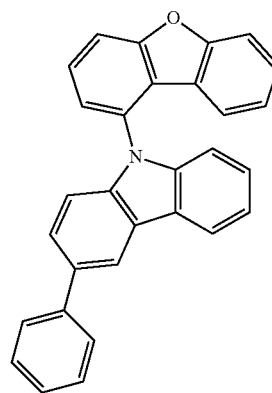
In the general formula (4), R⁷ to R⁹ each independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, an aromatic hydrocarbon group having 6 to 18 carbon atoms, or an aromatic heterocyclic group having 3 to 17 carbon atoms. However, in the case of the aromatic heterocyclic group having 3 to 17 carbon atoms, it is not preferred that the group be a nitrogen-containing six-membered ring.

Here, a substituted or unsubstituted aromatic heterocyclic group having 3 to 17 carbon atoms that is not a nitrogen-containing six-membered ring is preferably a substituted or unsubstituted, fused aromatic heterocyclic group having 6 to

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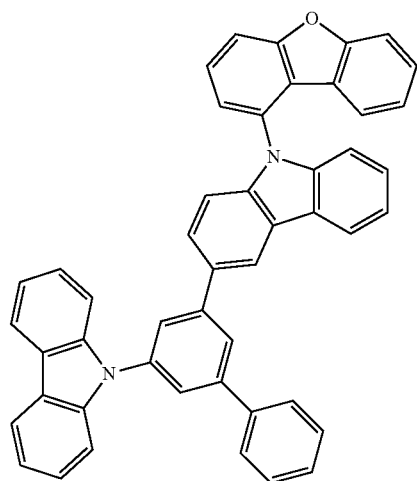
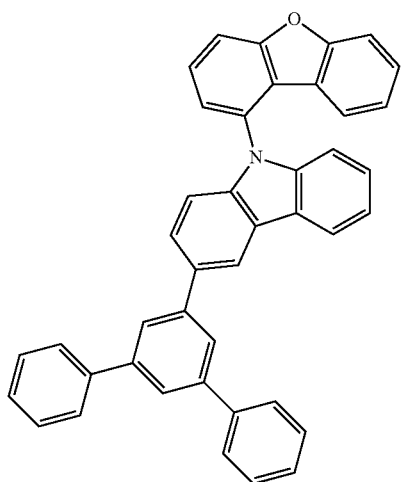
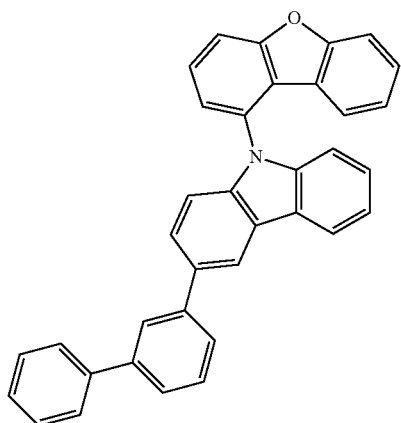
17 carbon atoms. Such aromatic heterocyclic group is the same as the aromatic heterocyclic group described above except that the group is not a nitrogen-containing six-membered ring.

Specific examples of the compound represented by the general formula (1) are shown below. However, the compound is not limited to the exemplified compounds.

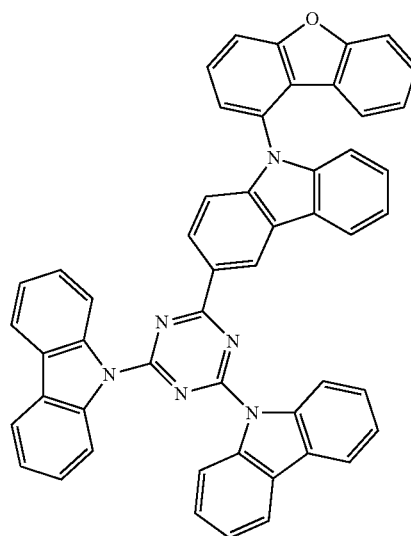
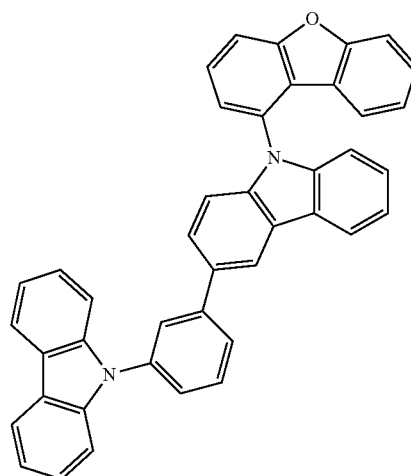
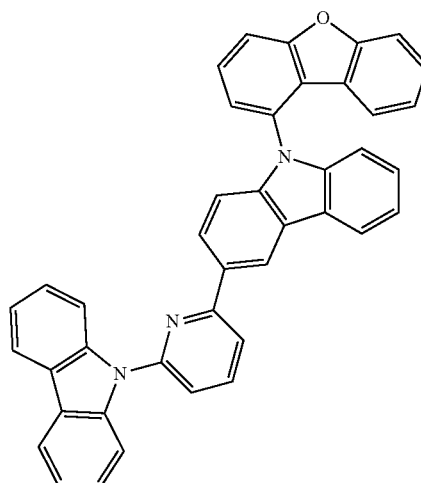


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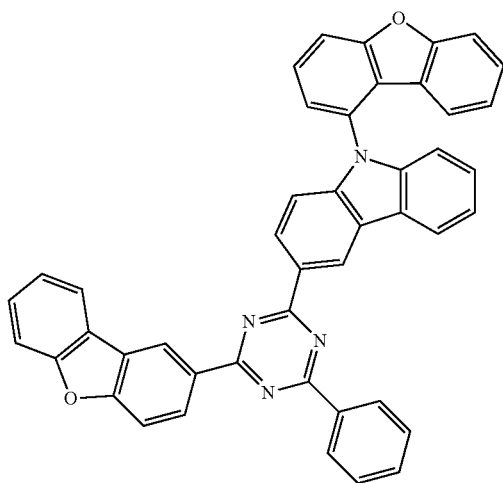
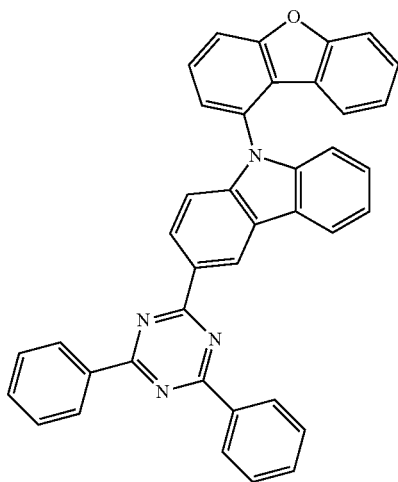
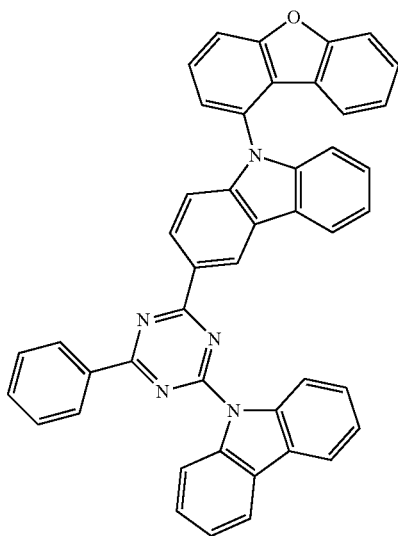
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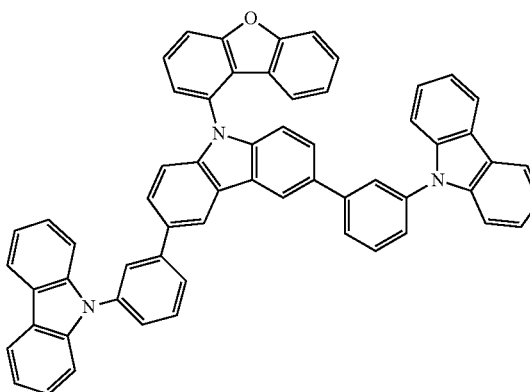
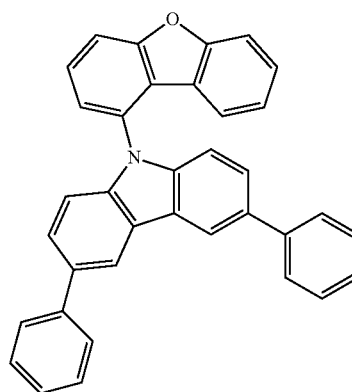
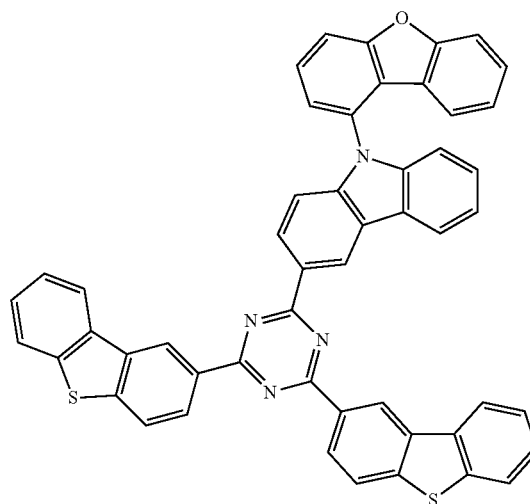
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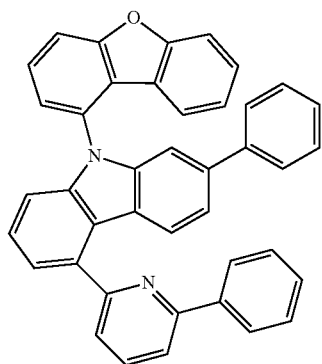
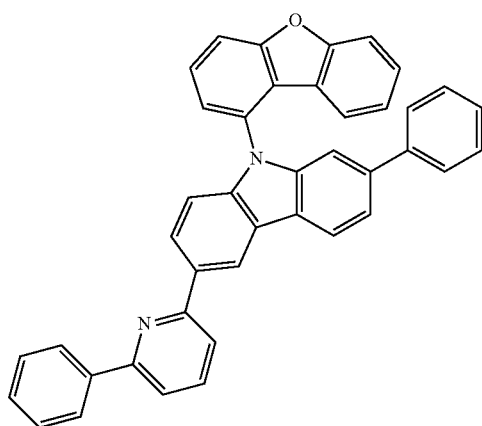
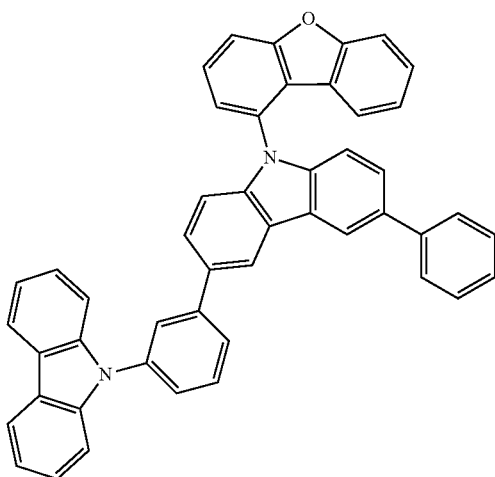
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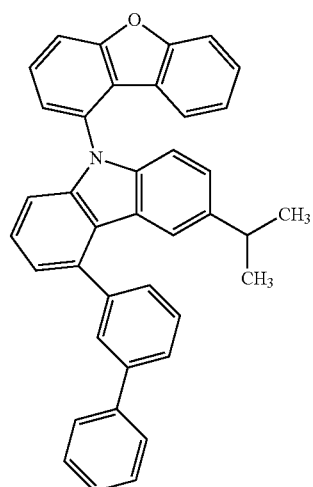
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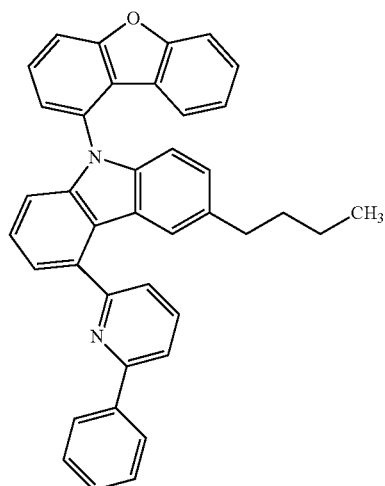
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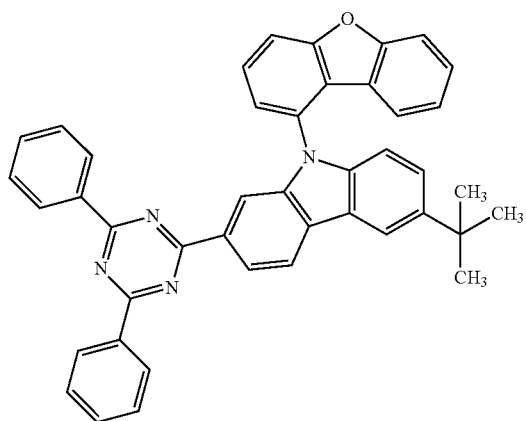
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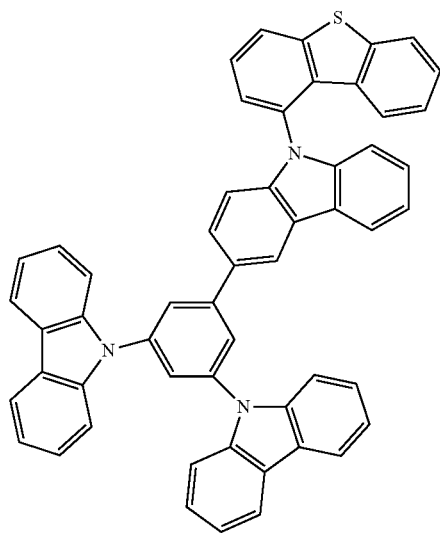
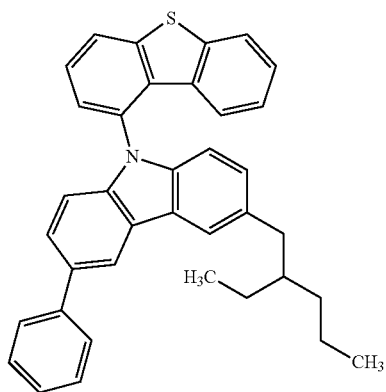
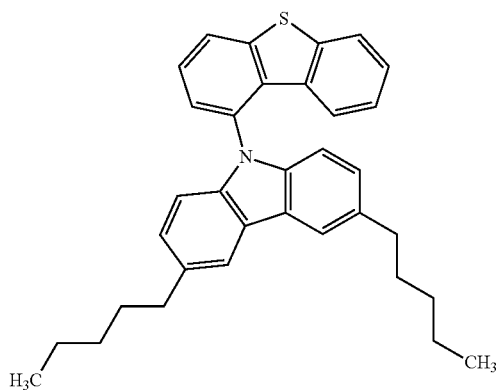
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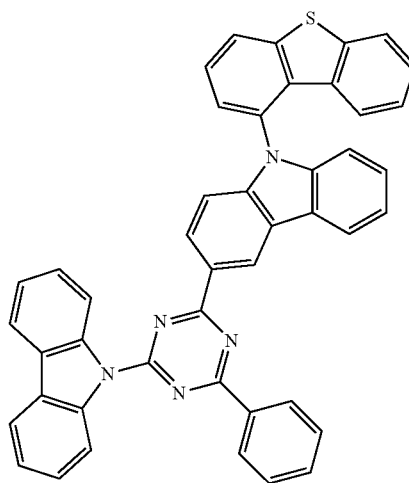
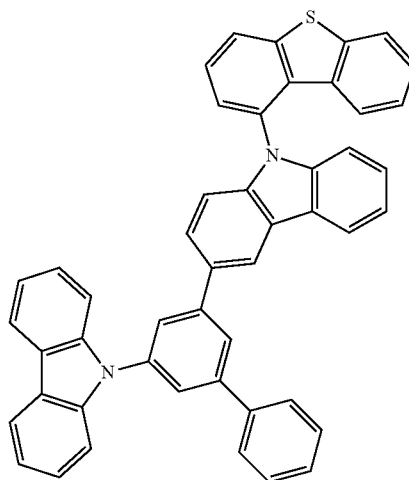
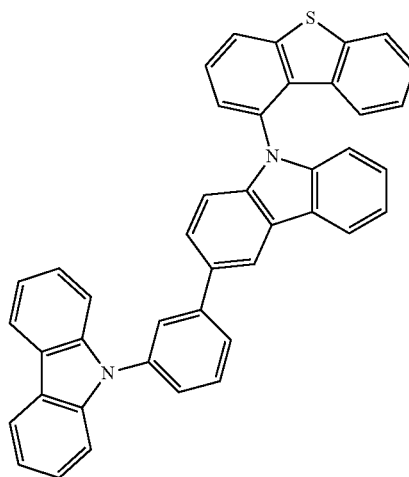
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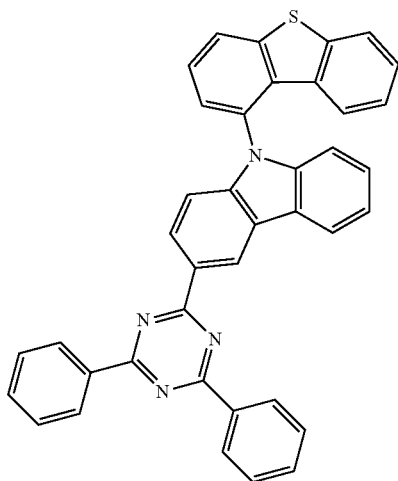
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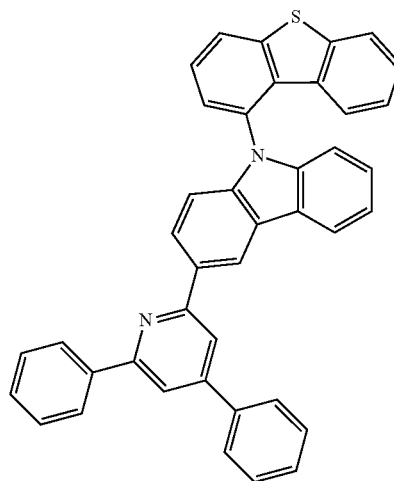
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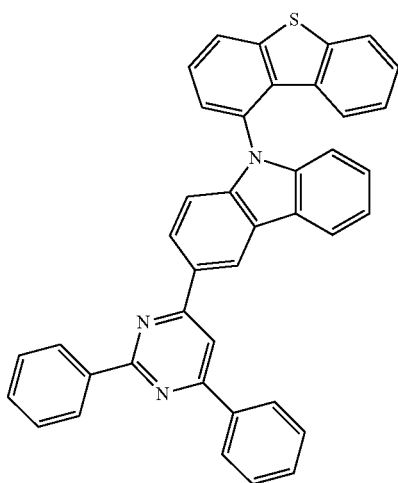
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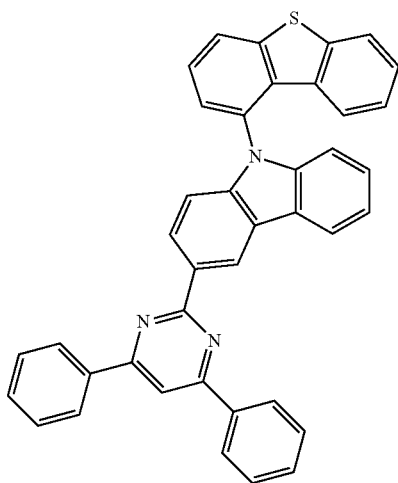
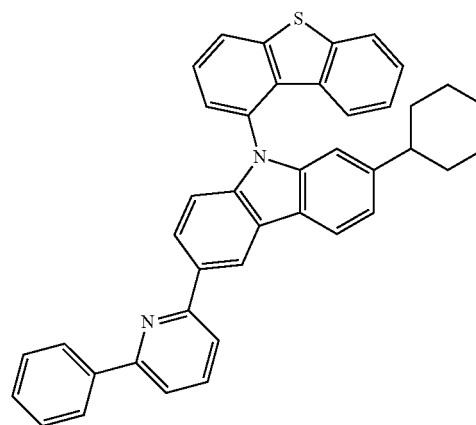
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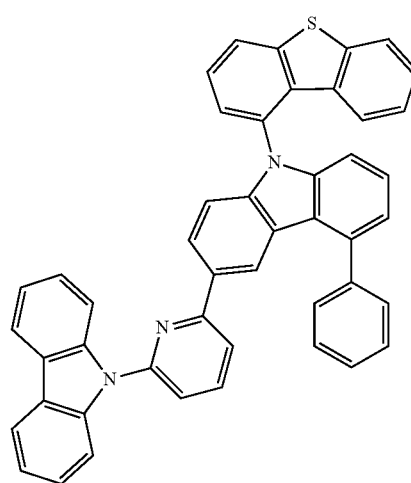
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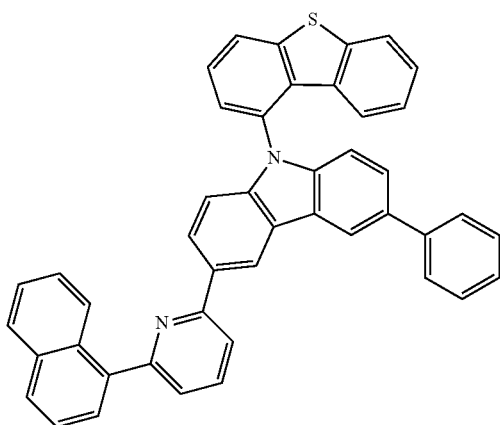


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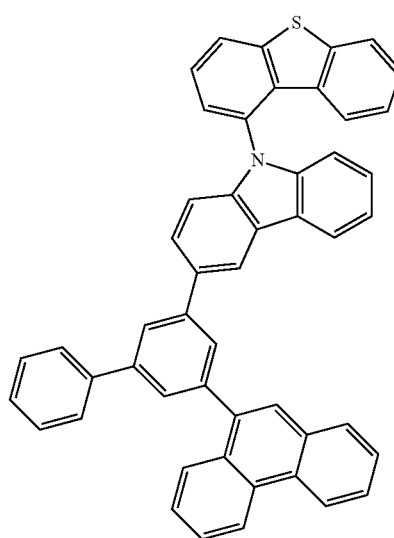
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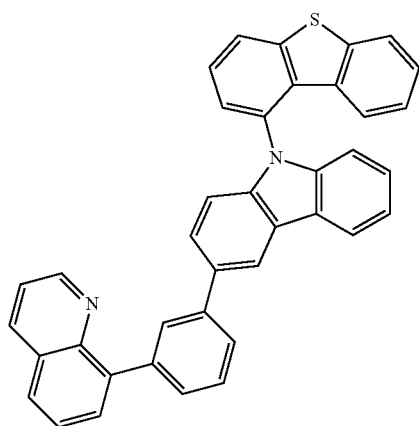
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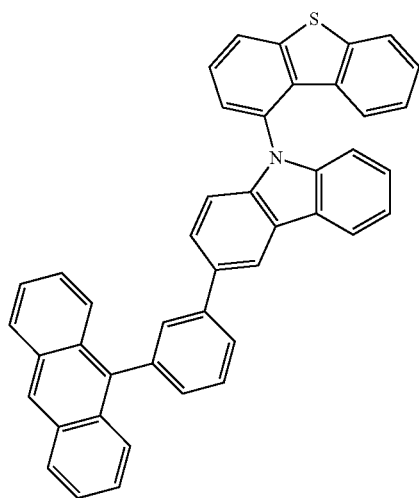
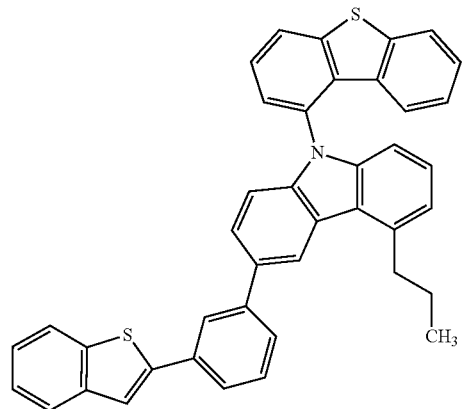
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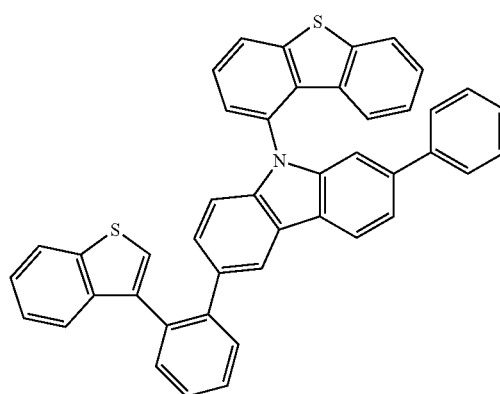
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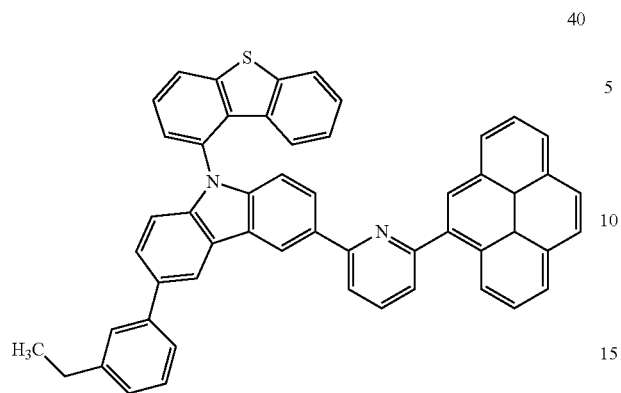


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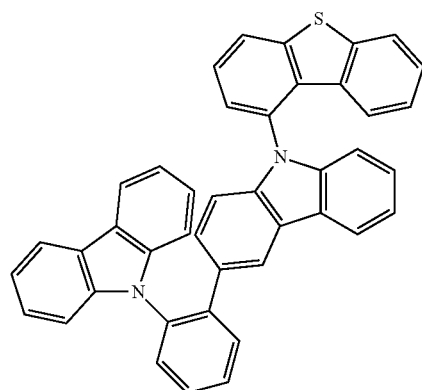
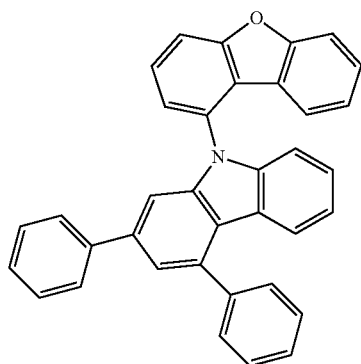
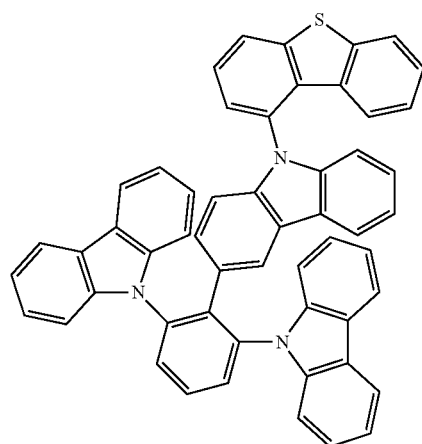
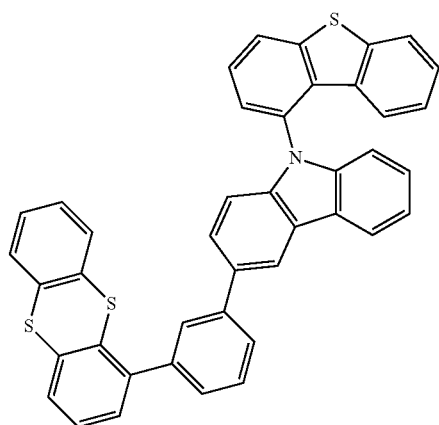
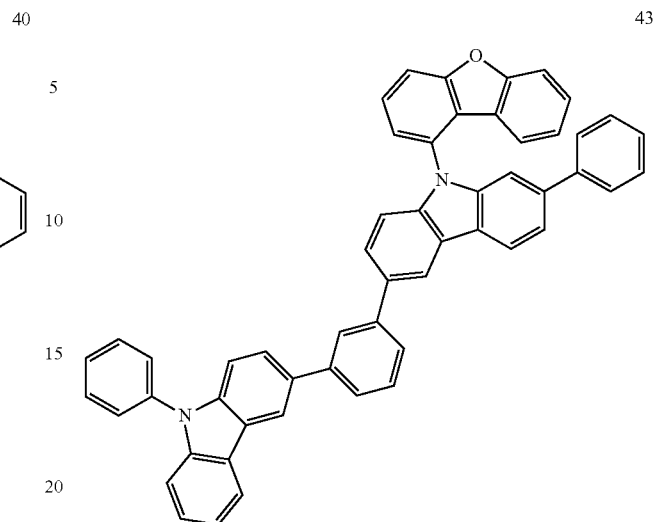


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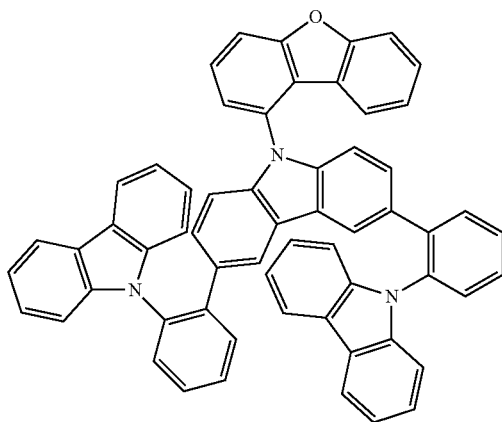
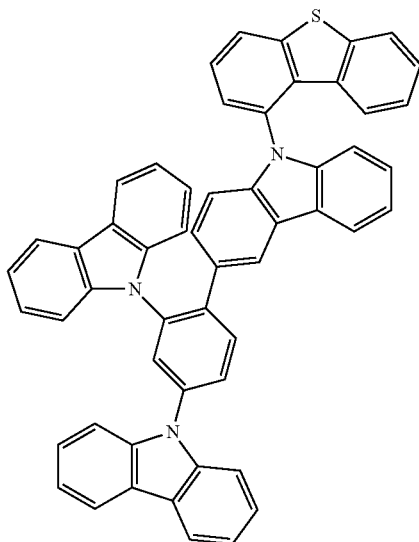
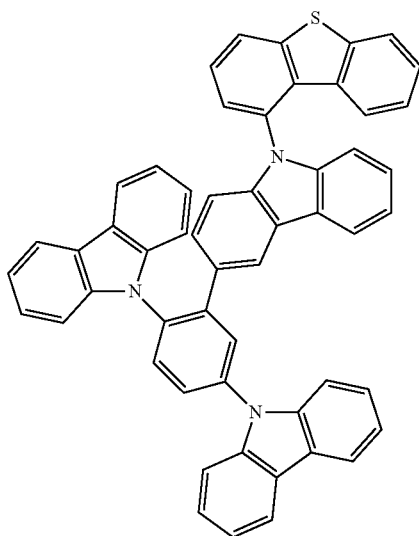
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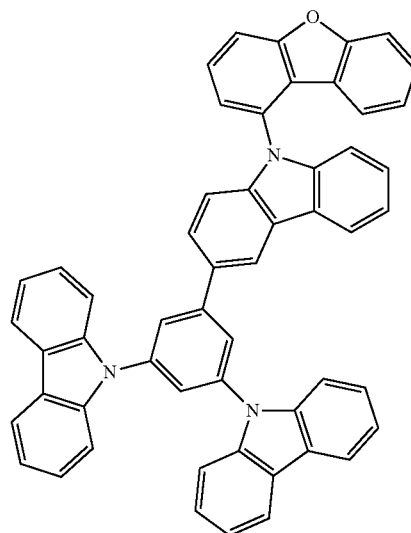
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When the carbazole compound represented by the general formula (1) is incorporated into at least one organic layer in an organic EL device formed by laminating an anode, a plurality of organic layers, and a cathode on a substrate, an excellent organic electroluminescent device is provided. The organic layers preferably include at least a light-emitting layer, and preferably further include a hole-transporting layer, an electron-transporting layer, a hole-blocking layer, or an electron-blocking layer. A light-emitting layer, a hole-transporting layer, an electron-transporting layer, a hole-blocking layer, or an electron-blocking layer is suitable as the organic layer into which the carbazole compound is incorporated. It is more preferred that the carbazole compound be incorporated as a host material in a light-emitting layer containing a phosphorescent light-emitting dopant.

The organic EL device of the present invention includes organic layers including at least one light-emitting layer between an anode and a cathode laminated on a substrate. In addition, at least one of the organic layers contains the carbazole compound. The carbazole compound represented by the general formula (1) is advantageously contained in the light-emitting layer together with a phosphorescent light-emitting dopant.

Next, the structure of the organic EL device of the present invention is described with reference to the drawings. However, the structure of the organic EL device of the present invention is by no means limited to one illustrated in the drawings.

FIG. 1 is a sectional view illustrating a structural example of a general organic EL device. Reference numerals 1, 2, 3, 4, 5, 6, and 7 represent a substrate, an anode, a hole-injecting layer, a hole-transporting layer, a light-emitting layer, an electron-transporting layer, and a cathode, respectively. The organic EL device of the present invention may include an exciton-blocking layer adjacent to the light-emitting layer, or may include an electron-blocking layer between the light-emitting layer and the hole-injecting layer. The exciton-blocking layer may be inserted on any of the anode side and the cathode side of the light-emitting layer, and may also be inserted simultaneously on both sides. The organic EL device of the present invention includes the substrate, the anode, the light-emitting layer, and the cathode as its essential layers. The organic EL device of the present invention

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preferably includes a hole-injecting/transporting layer and an electron-injecting/transporting layer in addition to the essential layers, and more preferably includes a hole-blocking layer between the light-emitting layer and the electron-injecting/transporting layer. It should be noted that the hole-injecting/transporting layer means any one or both of the hole-injecting layer and the hole-transporting layer, and that the electron-injecting/transporting layer means any one or both of an electron-injecting layer and the electron-transporting layer.

It should be noted that it is possible to adopt a reverse structure as compared to FIG. 1, that is, a structure formed by laminating the layers on the substrate 1 in the order of the cathode 7, the electron-transporting layer 6, the light-emitting layer 5, the hole-transporting layer 4, and the anode 2. In this case as well, a layer may be added or eliminated as required.

—Substrate—

The organic EL device of the present invention is preferably supported by a substrate. The substrate is not particularly limited, and any substrate that has long been conventionally used for an organic EL device may be used. For example, a substrate made of glass, a transparent plastic, quartz, or the like may be used.

—Anode—

Preferably used as the anode in the organic EL device is an anode formed by using, as an electrode substance, any of a metal, an alloy, an electrically conductive compound, and a mixture thereof, all of which have a large work function (4 eV or more). Specific examples of such electrode substance include metals such as Au and conductive transparent materials such as CuI, indium tin oxide (ITO), SnO_2 , and ZnO. Further, it may be possible to use a material such as IDIXO ($\text{In}_2\text{O}_3\text{—ZnO}$), which may be used for manufacturing an amorphous, transparent conductive film. In order to produce the anode, it may be possible to form any of those electrode substances into a thin film by using a method such as vapor deposition or sputtering and form a pattern having a desired shape thereon by photolithography. Alternatively, in the case of not requiring high pattern accuracy (about 100 μm or more), a pattern may be formed via a mask having a desired shape when any of the above-mentioned electrode substances is subjected to vapor deposition or sputtering. Alternatively, when a coatable substance such as an organic conductive compound is used, it is also possible to use a wet film-forming method such as a printing method or a coating method. When luminescence is taken out from the anode, the transmittance of the anode is desirably controlled to more than 10%. Further, the sheet resistance as the anode is preferably several hundred Ω/\square or less. Further, the thickness of the resultant film is, depending on the material used, selected from usually the range of 10 to 1,000 nm, preferably the range of 10 to 200 nm.

—Cathode—

On the other hand, used as the cathode is a cathode formed by using, as an electrode substance, any of a metal (referred to as electron-injecting metal), an alloy, an electrically conductive compound, and a mixture thereof, all of which have a small work function (4 eV or less). Specific examples of such electrode substance include sodium, a sodium-potassium alloy, magnesium, lithium, a magnesium/copper mixture, a magnesium/silver mixture, a magnesium/aluminum mixture, a magnesium/indium mixture, an aluminum/aluminum oxide (Al_2O_3) mixture, indium, a lithium/aluminum mixture, and a rare earth metal. Of those, for example, a mixture of an electron-injecting metal and a second metal as a stable metal having a larger work function value than the

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former metal, such as a magnesium/silver mixture, a magnesium/aluminum mixture, a magnesium/indium mixture, an aluminum/aluminum oxide (Al_2O_3) mixture, or a lithium/aluminum mixture, or aluminum is suitable from the viewpoints of electron-injecting property and durability against oxidation or the like. The cathode may be produced by forming any of those electrode substances into a thin film by using a method such as vapor deposition or sputtering. Further, the sheet resistance as the cathode is preferably several hundred Ω/\square or less, and the thickness of the resultant film is selected from usually the range of 10 nm to 5 μm , preferably the range of 50 to 200 nm. It should be noted that, in order for luminescence produced to pass through, any one of the anode and cathode of the organic EL device is preferably transparent or semi-transparent, because the light emission luminance improves.

Further, after any of the above-mentioned metals is formed into a film having a thickness of 1 to 20 nm as a cathode, any of the conductive transparent materials mentioned in the description of the anode is formed into a film on the cathode, thereby being able to produce a transparent or semi-transparent cathode. Then, by applying this, it is possible to produce a device in which both the anode and cathode have transparency.

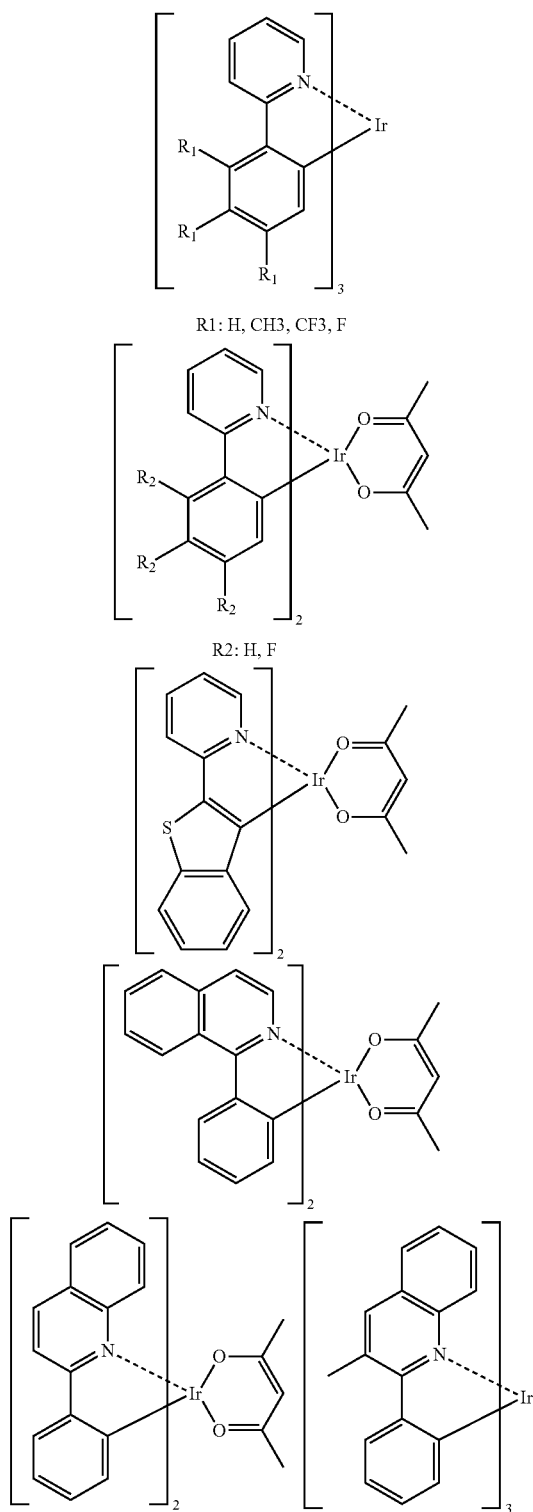
—Light-Emitting Layer—

The light-emitting layer is a phosphorescent light-emitting layer, and contains a phosphorescent light-emitting dopant and a host material. It is recommended to use, as a material for the phosphorescent light-emitting dopant, a material containing an organic metal complex including at least one metal selected from ruthenium, rhodium, palladium, silver, rhenium, osmium, iridium, platinum, and gold. Specific examples thereof include, but not limited to, the compounds disclosed in the following patent publications. The numbers of the patent publications are shown below.

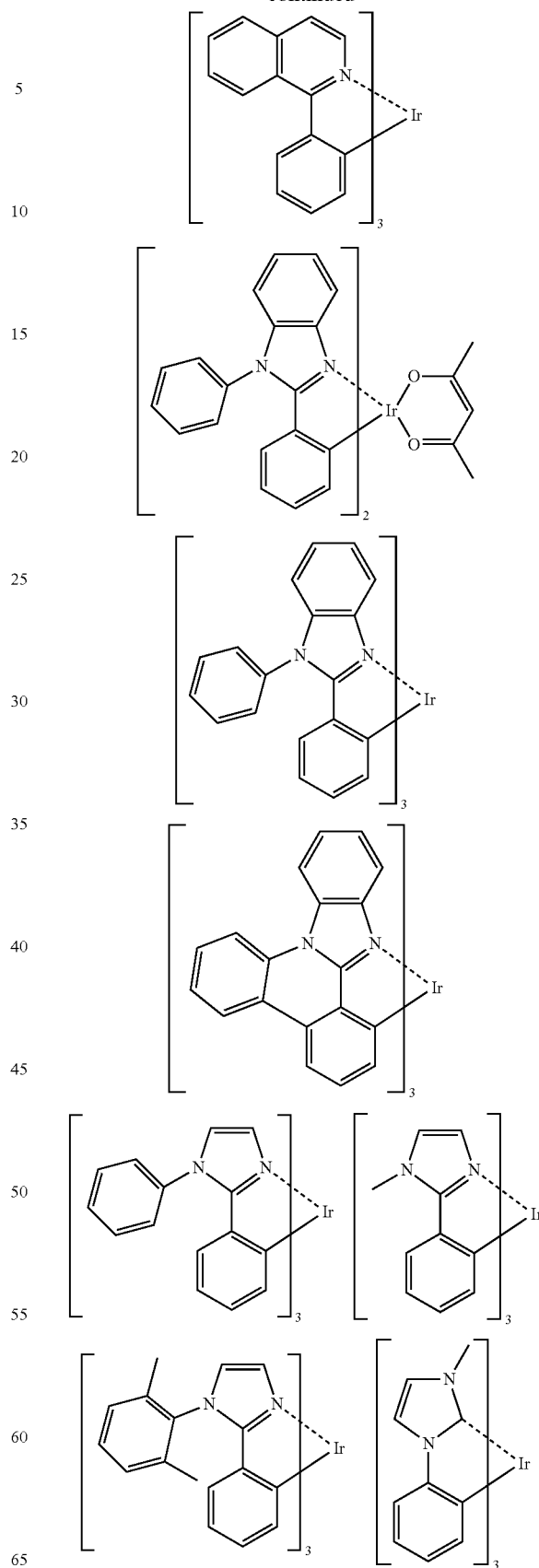
For example, WO 2009/073245 A1, WO 2009/046266 A1, WO 2007/095118 A3, WO 2008/156879 A1, WO 2008/140657 A1, US 2008/261076 A, JP 2008-542203 A, WO 2008/054584 A1, JP 2008-505925 A, JP 2007-522126 A, JP 2004-506305 A, JP 2006-513278 A, JP 2006-50596 A, WO 2006/046980 A1, WO 2005/113704 A3, US 2005/260449 A, US 2005/2260448 A, US 2005/214576 A, WO 2005/076380 A3, US 2005/119485 A, WO 2004/045001 A3, WO 2004/045000 A3, WO 2006/100888 A1, WO 2007/004380 A1, WO 2007/023659 A1, WO 2008/035664 A1, JP 2003-272861 A, JP 2004-111193 A, JP 2004-319438 A, JP 2007-2080 A, JP 2007-9009 A, JP 2007-227948 A, JP 2008-91906 A, JP 2008-311607 A, JP 2009-19121 A, JP 2009-46601A, JP 2009-114369A, JP2003-253128A, JP 2003-253129 A, JP 2003-253145 A, JP 2005-38847 A, JP 2005-82598 A, JP 2005-139185 A, JP 2005-187473 A, JP 2005-220136 A, JP2006-63080A, JP 2006-104201 A, JP2006-111623A, JP2006-213720A, JP2006-290891A, JP2006-298899 A, JP 2006-298900 A, WO 2007/018067 A1, WO 2007/058080 A1, WO 2007/058104 A1, JP 2006-131561 A, JP 2008-239565 A, JP 2008-266163 A, JP 2009-57367A, JP 2002-117978 A, JP 2003-123982 A, JP 2003-133074 A, JP 2006-93542A, JP 2006-131524 A, JP 2006-261623 A, JP 2006-303383 A, JP 2006-303394A, JP 2006-310479A, JP 2007-88105A, JP 2007-258550 A, JP 2007-324309A, JP 2008-270737A, JP 2009-96800A, JP 2009-161524 A, WO 2008/050733 A1, JP 2003-73387A, JP 2004-59433 A, JP 2004-155709 A, JP 2006-104132 A, JP 2008-37848A, JP 2008-133212 A, JP 2009-57304 A, JP 2009-286716A, JP 2010-83852 A, JP 2009-532546 A, JP 2009-536681 A, and JP 2009-542026 A.

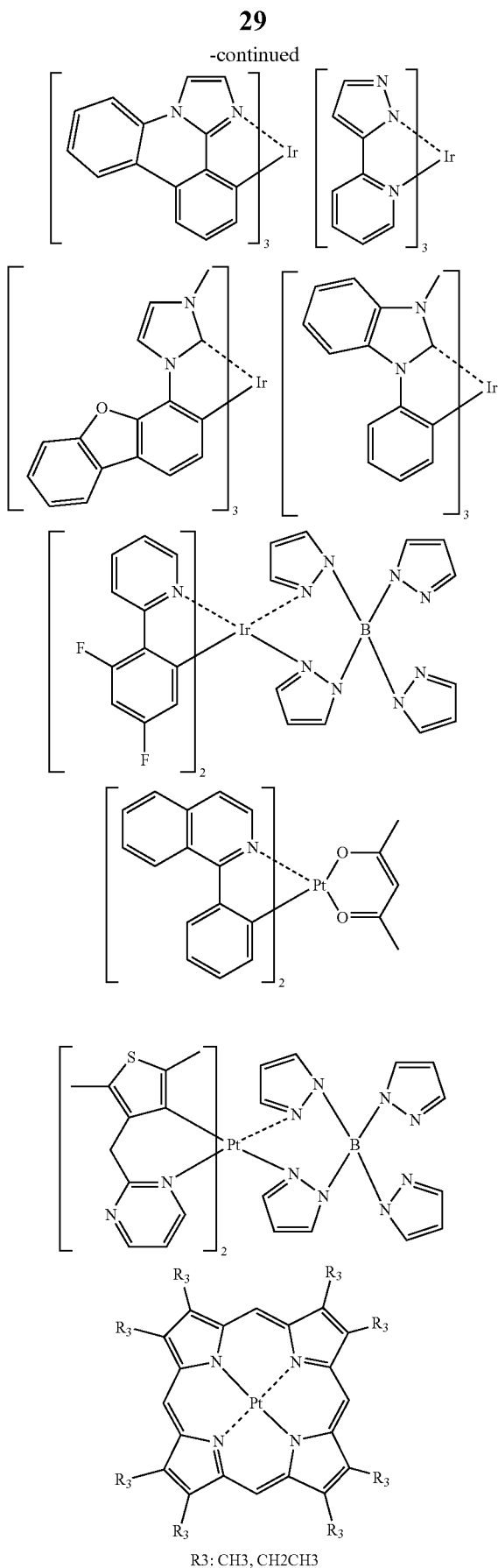
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Preferred examples of the phosphorescent light-emitting dopant include complexes such as Ir(ppy)₃, complexes such as Ir(Bt)2•acac₃, and complexes such as PtOEt₃, the complexes each having a noble metal element such as Ir as a central metal. Specific examples of those complexes are shown below, but the complexes are not limited to the compounds described below.

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It is preferred that the content of the phosphorescent light-emitting dopant in the light-emitting layer fall within the range of 0.1 to 50 wt %, more preferably 1 to 30 wt %.

It is preferred to use, as a host material in the light-emitting layer, the carbazole compound represented by the general formula (1). However, when the carbazole compound is used in any of the organic layers other than the light-emitting layer, the material to be used in the light-emitting layer may be any other host material other than the carbazole compound, and the carbazole compound and any other host material may be used in combination. Further, a plurality of kinds of known host materials may be used in combination.

It is preferred to use, as a usable known host compound, a compound that has a hole-transporting ability or an electron-transporting ability, is capable of preventing luminescence from having a longer wavelength, and has a high glass transition temperature.

Such other host materials are known because they are mentioned in many patent literatures and the like, and hence may be chosen from those in the patent literatures and the like. Specific examples of the host material include, but not particularly limited to, an indole derivative, a carbazole derivative, an indolocarbazole derivative, a triazole derivative, an oxazole derivative, an oxadiazole derivative, an imidazole derivative, a polyarylethane derivative, a pyrazoline derivative, a pyrazolone derivative, a phenylenediamine derivative, an arylamine derivative, an amino-substituted chalcone derivative, a styrylanthracene derivative, a fluorenone derivative, a hydrazone derivative, a stilbene derivative, a silazane derivative, an aromatic tertiary amine compound, a styrylamine compound, an aromatic dimethyldiene-based compound, a porphyrin-based compound, an anthraquinodimethane derivative, an anthrone derivative, a diphenylquinone derivative, a thiopyran dioxide derivative, a heterocyclic tetracarboxylic acid anhydride such as naphthalene perylene, a phthalocyanine derivative, various metal complexes typified by a metal complex of an 8-quinolinol derivative, a metal phthalocyanine, and metal complexes of benzoxazole and benzothiazole derivatives, and polymer compounds such as a polysilane-based compound, a poly(N-vinylcarbazole) derivative, an aniline-based copolymer, a thiophene oligomer, a polythiophene derivative, a polyphenylene derivative, a polyphenylenevinylene derivative, and a polyfluorene derivative.

—Injecting Layer—

The injecting layer refers to a layer formed between an electrode and an organic layer for the purposes of lowering

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a driving voltage and improving a light emission luminance, and includes a hole-injecting layer and an electron-injecting layer. The injecting layer may be interposed between the anode and the light-emitting layer or the hole-transporting layer, or may be interposed between the cathode and the light-emitting layer or the electron-transporting layer. The injecting layer may be formed as required.

—Hole-Blocking Layer—

The hole-blocking layer has, in a broad sense, the function of an electron-transporting layer, and is formed of a hole-blocking material that has a remarkably small ability to transport holes while having a function of transporting electrons, and hence the hole-blocking layer is capable of improving the probability of recombining an electron and a hole by blocking holes while transporting electrons.

The carbazole compound represented by the general formula (1) is preferably used in the hole-blocking layer. However, when the carbazole compound is used in any other organic layer, a known material for a hole-blocking layer may be used. In addition, it is possible to use, as a material for the hole-blocking layer, any of materials for the electron-transporting layer to be described later as required.

—Electron-Blocking Layer—

The electron-blocking layer is formed of a material that has a remarkably small ability to transport electrons while having a function of transporting holes, and hence the electron-blocking layer is capable of improving the probability of recombining an electron and a hole by blocking electrons while transporting holes.

Any of materials for the hole-transporting layer to be described later can be used as required as a material for the electron-blocking layer. The thickness of the electron-blocking layer is preferably 3 to 100 nm, more preferably 5 to 30 nm.

—Exciton-Blocking Layer—

The exciton-blocking layer refers to a layer used for blocking excitons produced by the recombination of a hole and an electron in the light-emitting layer from diffusing in charge-transporting layers. The insertion of this layer enables effective confinement of the excitons in the light-emitting layer, thereby being able to improve the luminous efficiency of the device. The exciton-blocking layer may be inserted on any of the anode side and the cathode side of the adjacent light-emitting layer, and may also be inserted simultaneously on both sides.

As a material for the exciton-blocking layer, there are given, for example, 1,3-dicarbazolylbenzene (mCP) and bis(2-methyl-8-quinolinolato)-4-phenylphenolatoaluminum (III) (BALq).

—Hole-Transporting Layer—

The hole-transporting layer is formed of a hole-transporting material having a function of transporting holes, and a single hole-transporting layer or a plurality of hole-transporting layers may be formed.

The hole-transporting material has any one of hole-injecting property, hole-transporting property, and electron-blocking property, and any of an organic compound and an inorganic compound may be used. It is preferred to use the carbazole compound represented by the general formula (1) in the hole-transporting layer. However, any compound selected from conventionally known compounds may be used. Examples of the known hole-transporting material that may be used include a triazole derivative, an oxadiazole derivative, an imidazole derivative, a polyaryllalkane derivative, a pyrazoline derivative and a pyrazolone derivative, a phenylenediamine derivative, an arylamine derivative, an amino-substituted chalcone derivative, an oxazole deriva-

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tive, a styrylanthracene derivative, a fluorenone derivative, a hydrazone derivative, a stilbene derivative, a silazane derivative, an aniline-based copolymer, and a conductive high-molecular weight oligomer, in particular, a thiophene oligomer. However, a triazole derivative, an oxadiazole derivative, an imidazole derivative, an arylamine derivative, or an oxazole derivative is preferably used, and an arylamine derivative is more preferably used.

—Electron-Transporting Layer—

The electron-transporting layer is formed of a material having a function of transporting electrons, and a single electron-transporting layer or a plurality of electron-transporting layers may be formed.

An electron-transporting material (which also serves as a hole-blocking material in some cases) has only to have a function of transferring electrons injected from the cathode into the light-emitting layer. Although the carbazole compound represented by the general formula (1) according to the present invention is preferably used in the electron-transporting layer, any compound selected from conventionally known compounds may be used. Examples thereof include a nitro-substituted fluorene derivative, a diphenylquinone derivative, a thiopyran dioxide derivative, a carbodiimide, a fluorenylidene methane derivative, anthraquinodimethane and an anthrone derivative, and an oxadiazole derivative. Further, it is also possible to use, as the electron-transporting material, a thiadiazole derivative prepared by substituting an oxygen atom on an oxadiazole ring with a sulfur atom in the oxadiazole derivative and a quinoxaline derivative that has a quinoxaline ring known as an electron withdrawing group. Further, it is also possible to use a polymer material in which any of those materials is introduced in a polymer chain or is used as a polymer main chain.

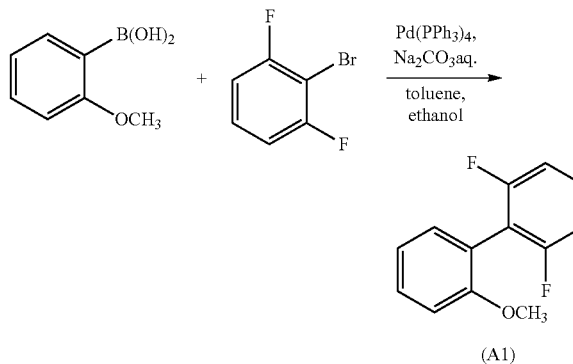
EXAMPLES

Hereinafter, the present invention is described in more detail by way of Examples. It should be appreciated that the present invention is not limited to Examples below and may be carried out in various forms as long as the various forms do not deviate from the gist of the present invention.

The routes described below were used to synthesize a carbazole compound to be used as a material for a phosphorescent light-emitting device. It should be noted that the number of each compound corresponds to the number given to the exemplified compound.

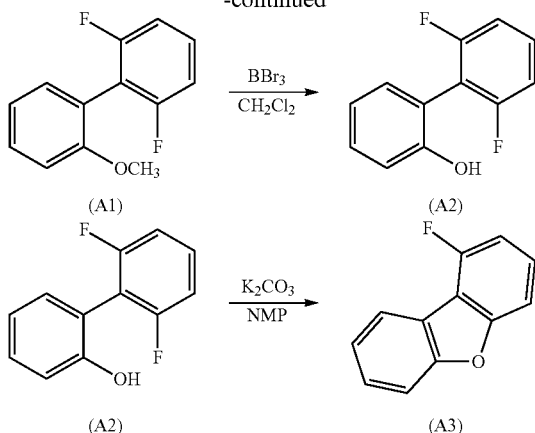
Example 1

Synthesis of (Compound 8)



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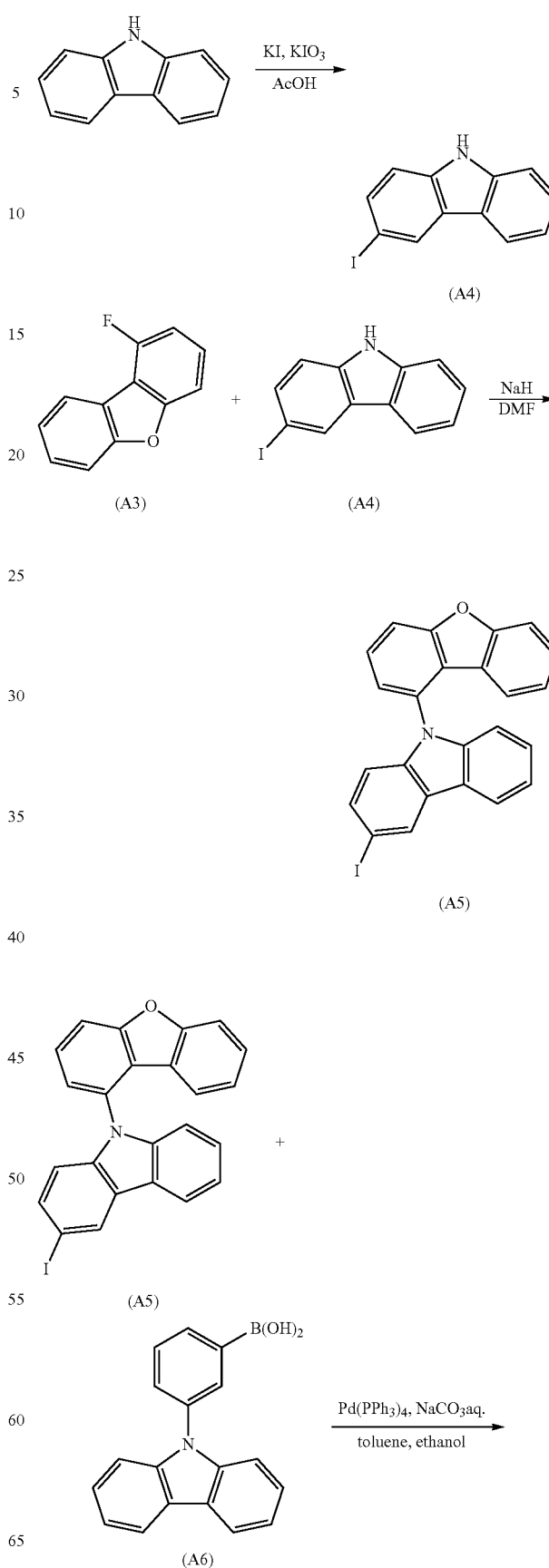


Under a nitrogen atmosphere, 18.84 g (0.124 mol) of 2-methoxyphenylboronic acid, 24.00 g (0.124 mol) of 1-bromo-2,6-difluorobenzene, 7.72 g (0.00496 mol) of tetrakis(triphenylphosphine)palladium(0), 600 ml of toluene, and 100 ml of ethanol were loaded, and then 200 ml of a 2 M aqueous solution of sodium hydroxide were added to the mixture while the mixture was stirred at room temperature. The resultant was stirred at 90° C. for 5 hr, and was then cooled to room temperature, followed by the washing of the organic layer with distilled water (300 ml×3). After the organic layer had been dried with anhydrous magnesium sulfate, magnesium sulfate was separated by filtration and then the solvent was distilled off under reduced pressure. The resultant residue was purified by silica gel column chromatography to provide 10.01 g (0.0455 mol, 38% yield) of an intermediate (A1) as a white solid.

Under a nitrogen atmosphere, 10.00 g (0.0454 mol) of the intermediate (1) and 100 ml of dichloromethane were loaded, and then 20 ml of a solution of boron tribromide in dichloromethane were added to the mixture while the mixture was stirred at 0° C. The resultant was stirred at room temperature for 6 hr, and then water was added thereto, followed by the washing of the organic layer with distilled water (30 ml×3). After the organic layer had been dried with anhydrous magnesium sulfate, magnesium sulfate was separated by filtration and then the solvent was distilled off under reduced pressure. The resultant residue was purified by silica gel column chromatography to provide 8.62 g (0.0418 mol, 92% yield) of an intermediate (A2) as a transparent liquid.

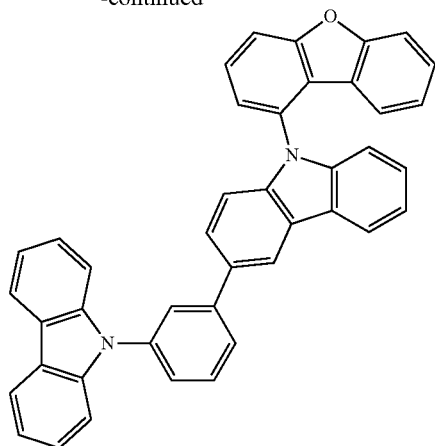
Under a nitrogen atmosphere, 8.62 g (0.0418 mol) of the intermediate (A2) and 230 ml of N-methyl-pyrrolidone were loaded, and then 11.56 g (0.0836 mol) of potassium carbonate were added to the mixture while the mixture was stirred at room temperature. The resultant was stirred at 180° C. for 3 hr, and was then cooled to room temperature, followed by the separation of potassium carbonate by filtration. 900 ml of distilled water were added to the filtrate and then the mixture was stirred at room temperature. After that, the precipitated solid was separated by filtration. The resultant solid was dissolved in dichloromethane and then the organic layer was washed with distilled water (30 ml×3). After the organic layer had been dried with anhydrous magnesium sulfate, magnesium sulfate was separated by filtration and then the solvent was distilled off under reduced pressure. The resultant residue was purified by silica gel column chromatography to provide 7.02 g (0.0377 mol, 90% yield) of an intermediate (A3) as a white solid.

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(8)

Under a nitrogen atmosphere, 35 g (0.209 mol) of carbazole and 300 ml of acetic acid were loaded, and then 24.24 g (0.146 mol) of potassium iodide and 31.24 g (0.146 mol) of potassium iodate were added to the mixture while the mixture was stirred at room temperature. The resultant was stirred at 80° C. for 2 hr, and was then cooled to room temperature. 300 ml of an aqueous solution of sodium hydrogen sulfite and 300 ml of tetrahydrofuran were added to the resultant, and then the mixture was stirred at room temperature. 300 ml of toluene were added to the resultant and then the organic layer was washed with distilled water (200 ml×2). After the organic layer had been dried with anhydrous magnesium sulfate, magnesium sulfate was separated by filtration and then the solvent was distilled off under reduced pressure. The resultant residue was purified by recrystallization to provide 20.75 g (0.0708 mol) of an intermediate (A4) as a white solid.

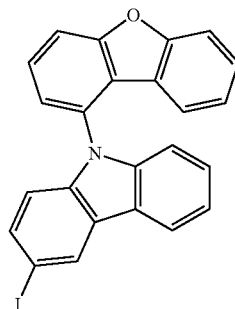
Under a nitrogen atmosphere, 3.03 g (0.0752 mol) of sodium hydride and 20 ml of dimethylformamide (DMF) were loaded, and then 20 ml of a DMF solution in which 20.00 g (0.0682 mol) of the intermediate (A4) had been dissolved were added to the mixture while the mixture was stirred at room temperature. The resultant was stirred at room temperature for 30 min, and then 20 ml of a DMF solution in which 7.00 g (0.0376 mol) of the intermediate (A3) had been dissolved were added to the resultant. The mixture was stirred at 120° C. for 7 hr, and was then cooled to room temperature. 300 ml of distilled water were added to the mixture and then the whole was stirred at room temperature. The precipitated solid was separated by filtration. The resultant solid was dissolved in tetrahydrofuran and then 200 ml of distilled water were added to the solution. The mixture was extracted with toluene (100 ml×3) and then the organic layer was dried with anhydrous magnesium sulfate. After that, magnesium sulfate was separated by filtration and then the solvent was distilled off under reduced pressure. The resultant residue was purified by silica gel column chromatography to provide 16.74 g (0.0364 mol, 97% yield) of an intermediate (A5) as a white solid.

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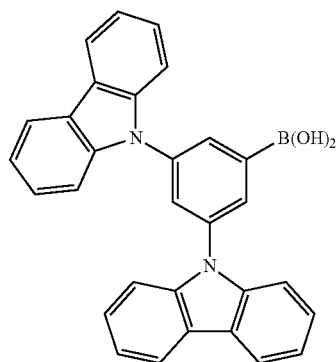
Under a nitrogen atmosphere, 10.00 g (0.0218 mol) of the intermediate (A5), 8.13 g (0.0283 mol) of the intermediate (A6), 1.36 g (0.000872 mol) of tetrakis(triphenylphosphine) palladium(0), 300 ml of toluene, and 50 ml of ethanol were loaded, and then 40 ml of a 2 M aqueous solution of sodium hydroxide were added to the mixture while the mixture was stirred at room temperature. The resultant was stirred at 90° C. for 2 hr, and was then cooled to room temperature, followed by the washing of the organic layer with distilled water (100 ml×2). After the organic layer had been dried with anhydrous magnesium sulfate, magnesium sulfate was separated by filtration and then the solvent was distilled off under reduced pressure. The resultant residue was purified by silica gel column chromatography and recrystallization to provide 8.70 g (0.0151 mol, 69% yield) of a (compound 8) as a white solid.

The APCI-TOFMS of the compound showed an [M+1] peak at an m/z of 575. FIG. 2 shows the results of its 1H-NMR measurement (measurement solvent: THF-d8).

Example 2



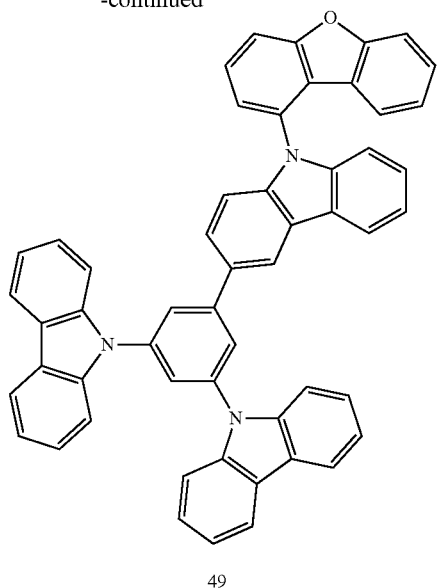
(A5)



(A7)

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Under a nitrogen atmosphere, 9.00 g (0.0196 mol) of the intermediate (A5), 10.63 g (0.0235 mol) of the intermediate (A7), 1.22 g (0.000784 mol) of tetrakis(triphenylphosphine) palladium(0), 300 ml of toluene, and 50 ml of ethanol were loaded, and then 35 ml of a 2 M aqueous solution of sodium hydroxide were added to the mixture while the mixture was stirred at room temperature. The resultant was stirred at 70° C. for 7 hr, and was then cooled to room temperature, followed by the washing of the organic layer with distilled water (100 ml×2). After the organic layer had been dried with anhydrous magnesium sulfate, magnesium sulfate was separated by filtration and then the solvent was distilled off under reduced pressure. The resultant residue was purified by silica gel column chromatography and recrystallization to provide 1.61 g (0.00218 mol, 9% yield) of a (compound 49) as a white solid.

The APCI-TOFMS of the compound showed an [M+1] peak at an m/z of 740. FIG. 3 shows the results of its 1H-NMR measurement (measurement solvent: THF-d8).

Example 3

Each thin film was laminated by a vacuum deposition method at a degree of vacuum of 4.0×10^{-5} Pa on a glass substrate on which an anode formed of an ITO substrate having a thickness of 110 nm had been formed. First, CuPC was formed into a layer having a thickness of 20 nm on the ITO. Next, NPB was formed into a layer having a thickness of 20 nm to serve as a hole-transporting layer. Next, the (compound 8) as a host material and Ir(ppy)₃ as a dopant were co-deposited from different deposition sources onto the hole-transporting layer to form a light-emitting layer having a thickness of 30 nm. At this time, the concentration of Ir(ppy)₃ was 10 wt %. Next, Alq3 was formed into a layer having a thickness of 40 nm to serve as an electron-transporting layer. Further, lithium fluoride (LiF) was formed into a layer having a thickness of 1 nm to serve as an electron-injecting layer on the electron-transporting layer. Finally, aluminum (Al) was formed into a layer having a thickness of 70 nm to serve as an electrode on the electron-injecting layer. Thus, an organic EL device was produced.

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An external power source was connected to the resultant organic EL device to apply a DC voltage to the device. As a result, it was confirmed that the device had such light-emitting characteristics as shown in Table 1.

Examples 4 to 11

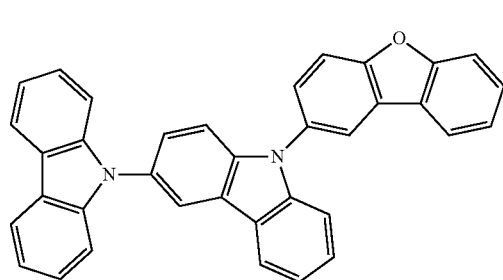
Organic EL devices were each produced in the same manner as in Example 3 except that compounds 1, 5, 6, 7, 24, 25, and 28 were synthesized in the same manner as in Example 1 and the compound 1, 5, 6, 7, 24, 25, or 28, or the compound 49 obtained in Example 2 was used instead of the compound 8 as the host material for the light-emitting layer in Example 3.

Comparative Example 1

An organic EL device was produced in the same manner as in Example 3 except that CEP was used as the host material for the light-emitting layer in Example 3.

Comparative Example 2

An organic EL device was produced in the same manner as in Example 3 except that a compound H-1 was used as the host material for the light-emitting layer in Example 3.



It was found that the local maximum wavelength of the emission spectrum of each of the devices obtained in Examples 3 to 11, and Comparative Examples 1 and 2 was 540 nm, and hence light emission from Ir(ppy)₃ was obtained. Table 1 shows the respective light-emitting characteristics. The columns "luminance", "voltage", and "luminous efficiency" in Table 1 show values (initial characteristics) at the time of driving at 20 mA/cm².

TABLE 1

	Host compound	Luminance (cd/m ²)	Voltage (V)	Visual luminous efficiency (lm/W)
Example 3	8	2360	9.4	3.9
4	1	2120	9.3	3.6
5	5	2130	9.0	3.7
6	6	2170	9.3	3.7
7	7	2060	9.4	3.4
8	24	2560	8.9	4.5
9	25	2290	9.3	3.9
10	28	1985	9.1	3.4
11	49	2230	9.4	3.7
Comparative Example 1	CBP	1120	8.7	2.0
2	H-1	1320	9.3	2.2

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Example 3 is improved in initial characteristics as compared to Comparative Example 1 and Comparative Example 2. The foregoing shows that the use of a compound, which has carbazole at the 1-position of dibenzothiophene or dibenzofuran and has a specific substituent at any one of the 2- to 7-positions of carbazole, in an organic EL device improves the characteristics of the organic EL device. The characteristics of the EL devices of Examples 4 to 11 are similarly good, which also shows the superiority of the carbazole compound represented by the general formula (1).

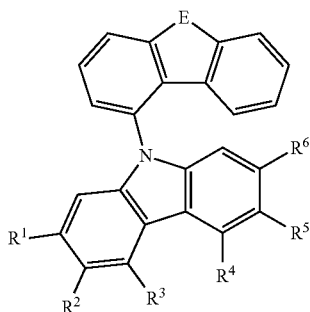
INDUSTRIAL APPLICABILITY

The carbazole compound represented by the general formula (1) to be used in the organic electroluminescent device of the present invention may enable the fine adjustment of hole and electron mobilities, and the control of various energy values, i.e., an ionization potential (IP), an electron affinity (EA), and a triplet energy (T1) because N of its carbazole ring is bonded to the 1-position of dibenzothiophene or dibenzofuran and the compound has a specific substituent at any one of the 2- to 7-positions of the carbazole ring. In addition, it may be possible to improve the stability of the carbazole compound in each of active states, i.e., oxidation, reduction, and excitation, and at the same time, the compound has a good amorphous characteristic. As a result of the foregoing, the compound can realize an organic EL device having a long driving lifetime and high durability.

The organic EL device according to the present invention has light-emitting characteristics, driving lifetime, and durability at practically satisfactory levels. Thus, the organic EL device has a high technical value in applications to flat panel displays (display devices for portable phones, in-vehicle display devices, display devices for OA computers, televisions, and the like), light sources utilizing characteristics of planar light emitters (light sources in lighting equipment and copying machines and backlight sources in liquid crystal displays and instruments), sign boards, sign lamps, and the like.

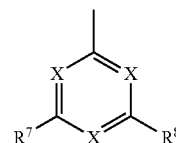
The invention claimed is:

1. An organic electroluminescent device, comprising:
a substrate;
an anode;
an organic layer; and
a cathode,
the anode, the organic layer, and the cathode being laminated on the substrate,
wherein the organic layer comprises at least one layer selected from the group consisting of a light-emitting layer, a hole-transporting layer, an electron-transporting layer, a hole-blocking layer, and an electron-blocking layer, the at least one layer containing a carbazole compound represented by the general formula (1):



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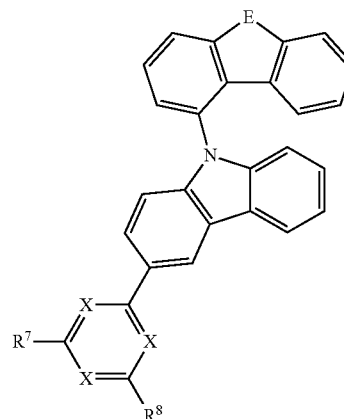
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in the general formula (1), R^1 to R^6 each independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, or an aromatic group represented by the general formula (2), and at least one of R^1 to R^6 represents an aromatic group represented by the general formula (2), X 's each independently represent CR^9 or nitrogen, R^7 to R^9 each independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, an aromatic hydrocarbon group having 6 to 18 carbon atoms, or an aromatic heterocyclic group having 3 to 17 carbon atoms, and the aromatic hydrocarbon group and the aromatic heterocyclic group may each have a substituent, and E represents oxygen or sulfur.

2. An organic electroluminescent device according to claim 1, wherein R^2 in the general formula (1) represents an aromatic group represented by the general formula (2).

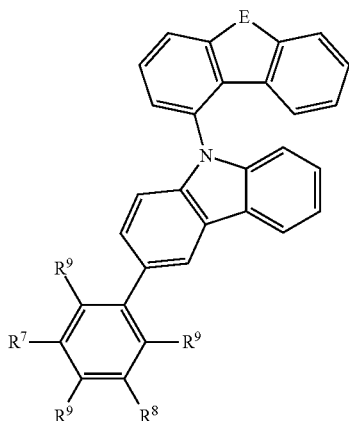
3. An organic electroluminescent device according to claim 1, wherein the carbazole compound comprises a carbazole compound represented by the general formula (3):



in the general formula (3), X , R^7 , R^8 , and E are identical in meaning to X , R^7 , R^8 , and E in the general formulae (1) and (2).

4. An organic electroluminescent device according to claim 3, wherein the carbazole compound comprises a carbazole compound represented by the general formula (4):

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in the general formula (4), R^7 , R^8 , and E are identical in meaning to R^7 , R^8 , and E in the general formula (3), and R^9 is identical in meaning to R^7 and a plurality of R^9 's may be identical to or different from one another.

5. An organic electroluminescent device according to claim 4, wherein R^7 to R^9 in the general formula (4) each

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- (4) independently represent hydrogen, an alkyl group having 1 to 10 carbon atoms, a cycloalkyl group having 3 to 11 carbon atoms, an unsubstituted aromatic hydrocarbon group having 6 to 18 carbon atoms, an unsubstituted aromatic heterocyclic group having 3 to 17 carbon atoms, or a substituted aromatic hydrocarbon group or a substituted aromatic heterocyclic group obtained by providing the unsubstituted aromatic hydrocarbon group or the unsubstituted aromatic heterocyclic group with a substituent, and the substituent comprises one kind selected from an alkyl group having 1 to 6 carbon atoms, a cycloalkyl group having 3 to 10 carbon atoms, an alkoxy group having 1 to 6 carbon atoms, an acyl group having 2 to 7 carbon atoms, an aromatic hydrocarbon group having 6 to 12 carbon atoms, and a diarylamino group having 12 to 24 carbon atoms.

6. An organic electroluminescent device according to claim 1, wherein the layer containing the carbazole compound comprises a light-emitting layer containing a phosphorescent light-emitting dopant.

7. An organic electroluminescent device according to claim 3, wherein the layer containing the carbazole compound comprises a light-emitting layer containing a phosphorescent light-emitting dopant.

* * * * *